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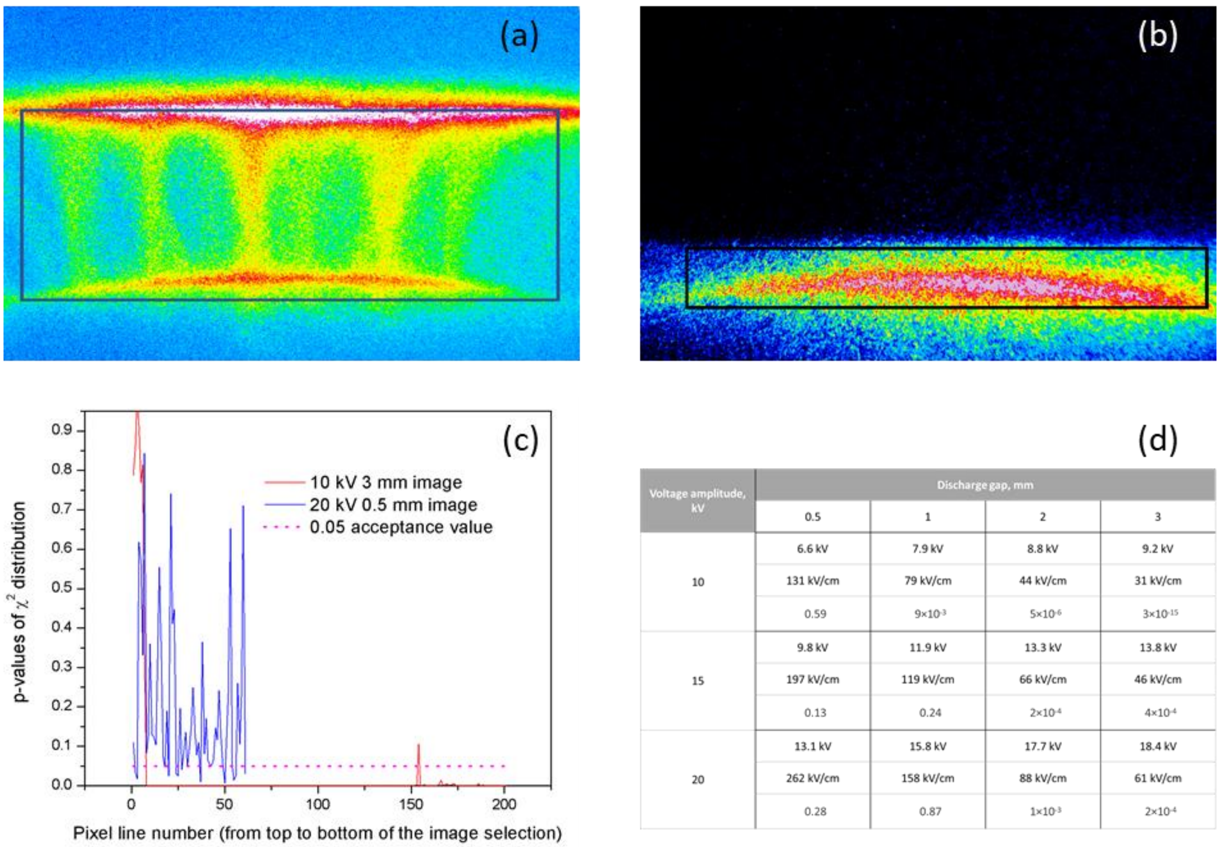
Dielectric barrier discharge (DBD), along with various types of plasma jets, has been extensively studied as a therapeutic tool for medical applications. DBD plasma is typically ignited in a direct contact with treated surfaces (e.g., skin) that play the role of a second electrode, providing a wide range of plasma effects – radiation (UV), neutral and charged species, and electric fields. In the case of nanosecond-pulsed high voltage plasma excitation regime, air DBD can be ignited in what appears to be a 'uniform' or diffuse mode, which is characterized by absence of streamer formation. Presence of streamers, which later become filaments at longer voltage pulse durations, results not only in non-uniform treatment of biological target but is also suggested to affect plasma chemistry due to higher local electric fields in the discharge gap. For example, it was shown that at the same treatment doses, non-uniform nanosecond-pulsed air DBD causes greater cell death rates compared to the uniform mode air DBD plasma, and one possible explanation is related to different concentrations of reactive species generated in these plasma regimes. One of the major components of plasmas that affects biological targets, in both DBD and plasma jet systems, which can be used as an indicator of plasma-induced cell toxicity, is hydrogen peroxide – a cytotoxic agent that is typically produced from short-living hydroxyl radicals.

In this work we studied OH radical generation in nanosecond dielectric barrier discharge in humid He/air gas. The measurements were carried out by means of laser-induced fluorescence (LIF) method at the PPPL Low Temperature Plasma Research Facility, directed by Dr. Shurik Yatom. Application of LIF towards OH density measurements is a widely used approach in plasmas generated in humid gas, particularly when temporal evolution of OH density is of interest. Here we examined the OH densities generated in the discharge ignited by application of nanosecond high-voltage pulses with amplitudes varying between 10 to 20 kV and interelectrode gap length varying between 0.5-3 mm. The peak voltage and the gap length determine the mode in which discharge is generated: homogenous or filamentary. These modes were previously identified and studied in an earlier publication. The measurements of OH in the gas phase were accompanied by the measurements of hydrogen peroxide (H_2O_2) in liquid water, downstream of the plasma and the relationship between these two were examined.

Here we performed an experimental study of nanosecond-pulsed DBD ignited in atmospheric pressure He-air atmosphere in presence of water. We show that the discharge, like in atmospheric air, operates in two distinctly different regimes: uniform and non-uniform (Figure 1). With the increase of the discharge gap from 0.5-1 mm to 2-3 mm and amplitudes of the high voltage pulses of 10-20 kV we observed discharge mode transition visually characterized by appearance of individual streamers, typical for long-pulsed bioelectric barrier plasma. Images of the discharge and their analysis using χ^2 statistical test show that the mode transition from streamer to diffuse mode happen when applied electric field approaches ~90 kV cm^{-1}, which is lower than for atmospheric air DBD (about 130 kV cm^{-1}).



Voltage amplitude, kV	Discharge gap, mm			
	0.5	1	2	3
10	6.6 kV	7.9 kV	8.8 kV	9.2 kV
	131 kV/cm	79 kV/cm	44 kV/cm	31 kV/cm
	0.59	9×10^{-3}	5×10^{-6}	3×10^{-15}
15	9.8 kV	11.9 kV	13.3 kV	13.8 kV
	197 kV/cm	119 kV/cm	66 kV/cm	46 kV/cm
	0.13	0.24	2×10^{-4}	4×10^{-4}
20	13.1 kV	15.8 kV	17.7 kV	18.4 kV
	262 kV/cm	158 kV/cm	88 kV/cm	61 kV/cm
	0.28	0.87	1×10^{-3}	2×10^{-4}

Figure 1. Single shot images (50 ns exposure) of the dielectric barrier discharge in non-uniform (a) and uniform (b) modes ignited at 3- and 0.5-mm gap distances and 10 kV and 20 kV voltage pulse amplitudes respectively. (c) Corresponding p-values of χ^2 distribution of pixels in the discharge images. (d) Calculated applied gap voltage, applied electric field values and corresponding average χ^2 distribution p-values for various discharge ignition conditions (discharge gap and peak voltage pulse values).

Based on the previously demonstrated much lower reduced electric fields in the uniform discharge operating in air compared to non-uniform plasma, we hypothesized that similar effect in He/air DBD would result in significantly different chemistries for uniform and non-uniform plasmas. We performed experimental measurements of gas-phase production of OH radicals in He/air DBD (Figure 2) and delivery of these radicals into liquid in the form of hydrogen peroxide, correlating them to the discharge uniformity modes.

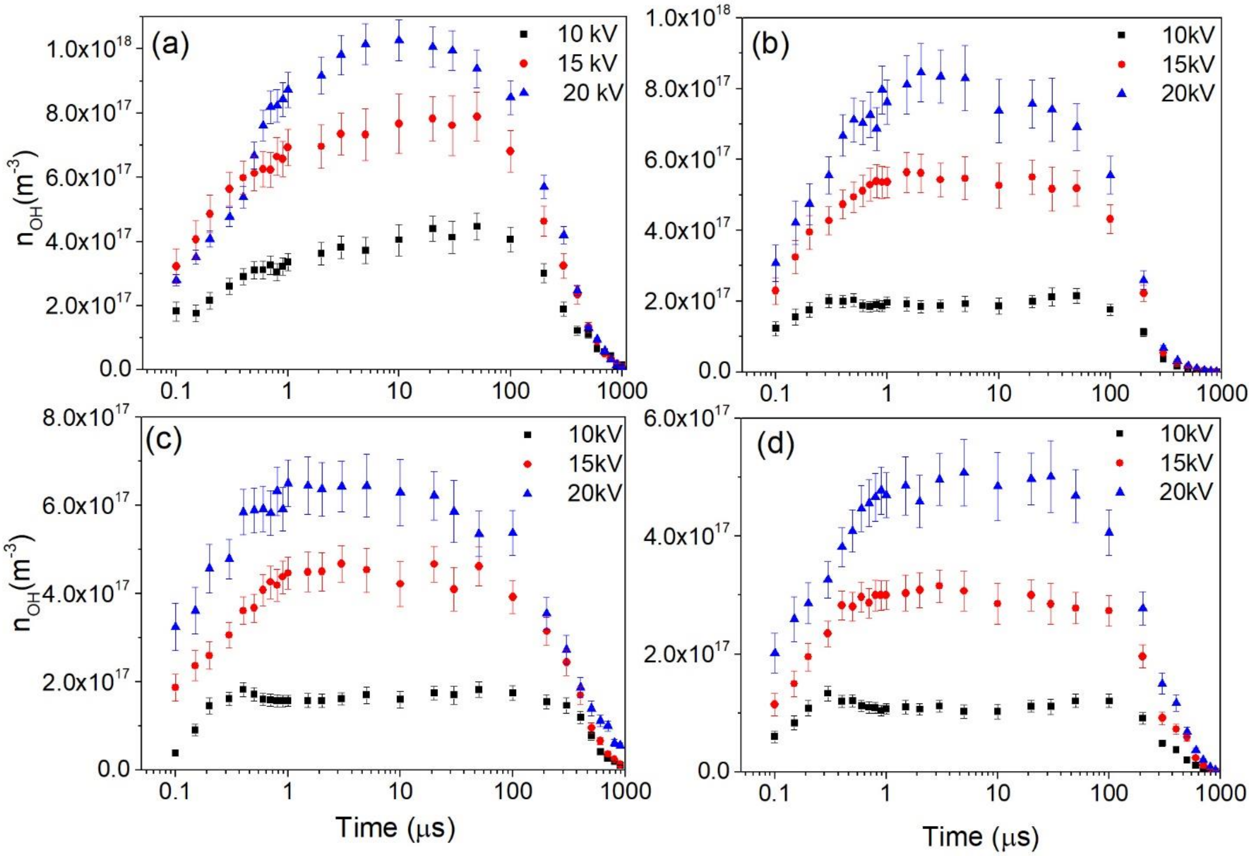


Figure 2. Temporal evolution of OH density for different cases of peak voltage amplitude 10-15-20 kV and different gaps (a) 0.5 mm (b) 1 mm (c) 2 mm (d) 3 mm.

The results show that, as expected, OH production in uniform mode is about 1.5 times less efficient than for streamer discharge: 38 nM/J vs 55 nM/J respectively (Figure 3). This effect is attributed to the decreased production of atomic oxygen in the absence of streamers (and, correspondingly, lower electric fields in the discharge). Simultaneously, we measure about 3 times lower efficiency of H_2O_2 delivery into liquid water of about 0.5 nM/J vs 1.7 nM/J for uniform and non-uniform DBD respectively – which corresponds to further decreased OH recombination of hydroxyl radicals in the case of atomic oxygen deficiency, resulting in lower production of hydrogen peroxide.

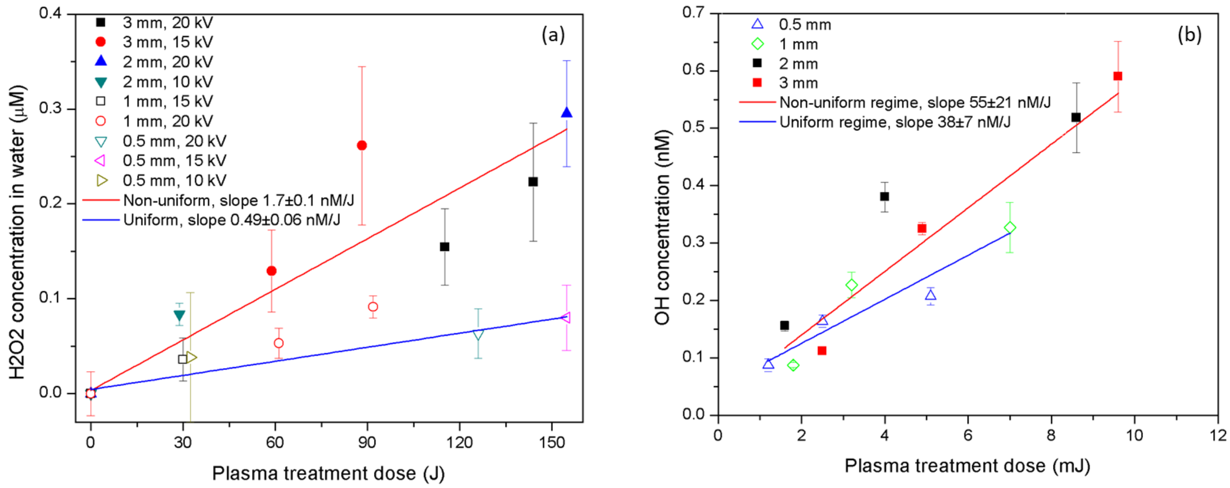


Figure 3. (a) Measured hydrogen peroxide concentrations in water as a function of treatment dose for various discharge regimes. (b) OH molar concentrations in gas phase.

The results of the study are being prepared for submission for a peer-reviewed journal publication, as well as to be presented at major international plasma conferences.